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## **THERMALLY INDUCED PERMEABILITY REDUCTION DUE TO PARTICLE MIGRATION IN SANDSTONES: THE EFFECT OF TEMPERATURE ON KAOLINITE MOBILISATION AND AGGREGATION.**

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### **ABSTRACT**

The seasonal imbalance in supply and demand of renewable energy requires seasonal storage, which potentially may be achieved by hot water injection in geothermal aquifers to minimize heat loss by advection. A reduction of porosity and permeability is a risk of heating the rock above the in-situ temperature. Published data indicate that the permeability reduction can be a consequence of the mobilisation of kaolinite particles. Particle mobilisation as a result of changes in physico-chemical conditions is often addressed using the DLVO theory (Derjaguin and Landau, 1941; Verwey and Overbeek, 1948). Mobilisation occurs due to similar surface charges on kaolinite and quartz grains which causes a detachment force on the kaolinite particles. Permeability reduction as a result of mobilisation requires the released particles to be captured in the pore throats. Data indicate that the permeability reduction can be reversible, implying captured particles are remobilised when the temperature is reduced.

This paper considers the effects of mineralogy and hydrodynamic forces on particle mobilisation and aggregation. The mineral surface charge originates from the interaction between the particles and the saturating fluid and is affected by both the fluid composition and the temperature. Kaolinite particles have a heterogeneous surface charge distribution that can play an important role in the particle aggregation in the pore throats, leading to permeability damage. The reduction of temperature causes an increase in the hydrodynamic force on the aggregated particles at pore throats. It can remobilise particles and lead to permeability recovery.

We discuss the mineral structure of quartz and kaolinite and estimate the effects of heating on the surface charges using published data. The DLVO theory is used to model the interaction energy between quartz and kaolinite particles for different

saturating fluids. The results are compared to the published data addressing the effect of temperature on permeability. This provides a qualitative explanation for the observed changes in permeability with temperature for the tests with distilled water.

### **INTRODUCTION**

Renewable energy production results in excess heat production during the summer in Denmark. This can be used in the municipal district heating system in winter, provided seasonal storage is available. Injection of heated water into sandstone aquifers that are currently exploited for geothermal energy is a promising and potential storage method. The in-situ temperatures of 60-100°C reduce heat loss from the water that is injected at temperatures in the range of 150-200°C.

Permeability reduction during hot water injection in sandstones is reported in some cases. Possible causes include: mineral precipitation; pore volume reduction due to thermal expansion of the solid fraction or compaction due to an increase in the pore volume compressibility; clogging by colloidal contamination and mobilisation of clay particles. Experimental data often show a recovery of the permeability when the temperature is reduced, (Weinbrandt and Ramey, 1975; Aruna, 1976; Casse and Ramey, 1979; Baudracco and Aoubouazza, 1995). An overview of published data by Rosenbrand and Fabricius (2012) indicates that the presence of kaolinite is the common factor in the experiments where permeability reduction is observed; when no kaolinite is present no permeability reduction is observed. However, there are tests where no permeability reduction occurs and kaolinite is present. This includes tests performed with distilled water and those with solutions with dissolved KCl or NaCl.

The mobilisation of kaolinite particles may not affect the porosity but the transport of kaolinite particles from the surface of the grains to the pore throats, as

illustrated in Figure 1, can cause a significant permeability reduction.

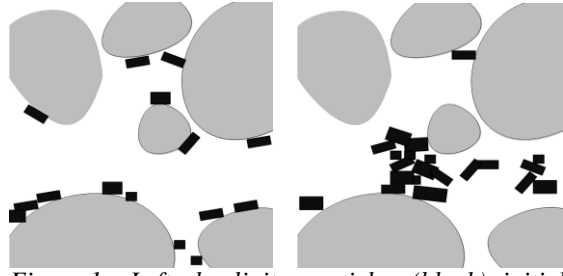


Figure 1: Left: kaolinite particles (black) initially present on the surface of quartz grains (gray). Right: mobilized particles aggregate and bridge pore throats.

Kaolinite mobilisation due to the change in temperature, ionic strength, or pH can be predicted using the DLVO theory (Khilar and Folger, 1984; Schembre and Kovsky, 2005). The DLVO theory accounts for the interaction of van der Waals attraction and the electrical double layer (EDL) force between the charged quartz and kaolinite surfaces. The sign and the order of magnitude of the surface charge depend on interaction with water molecules that adsorb on the mineral surface. At pH 6 both quartz and kaolinite have negative charges which attract oppositely charged ions that form a diffuse layer on the surface. Together the adsorbed and diffuse layers compose what is known as the electrical double layer (EDL). The EDL force results from the interaction of the double layers of two particles and is repulsive for similarly charged particles. It can result in repulsion between the particles when the EDL force exceeds the van der Waals force.

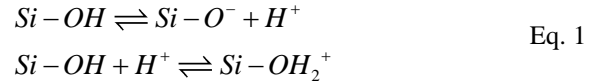
The kaolinite mineral consists of two different sheets that are bonded together (Grim, 1953). As a result, the surface charge is not uniformly distributed. In the solutions with a low pH, different sides of the mineral may have an opposite sign, resulting in flocculation of kaolinite suspensions (Wang and Siu, 2006).

When kaolinite particles are attached to the grain surface (Figure 1), heating can cause an increase of the EDL force between kaolinite and quartz resulting in the mobilisation of particles. Interaction between the suspended particles may cause the formation of a large aggregate that bridges the pore throat (Figure 1). The stability of the bridges is reduced at higher flow velocities due to larger hydrodynamic forces (Sen and Khilar, 2006). At constant flow velocity, the hydrodynamic forces may also be increased as a function of the fluid viscosity which increases when temperature is reduced. Together with the reduced repulsion from the quartz surface, the above mechanism can be a reason that cooling can recover the permeability.

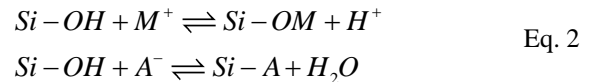
To understand the effects of the temperature on the mobilisation and the aggregation of particles, we consider the nature of the surface charge on kaolinite and quartz, and use the DLVO theory to estimate the effects of temperature on the EDL interaction between particles. Some of the published experimental results using different solutions and flow rates are compared to examine whether the observed permeability changes can be explained in the context of particle mobilisation and bridging. Due to the large number of parameters that affect these forces and the uncertainties of quantifications, the available data are insufficient to verify this mechanism.

### SURFACE CHARGE ON QUARTZ AND KAOLINITE

Quartz ( $\text{SiO}_2$ ) is built up of silica tetrahedra that consist of one silicon atom bonded to 4 oxygen atoms. Each oxygen atom is shared by two tetrahedra. On the mineral surface bonds are broken, which results in reactive sites. These interact with water molecules to form silanol groups that gain or lose  $\text{H}^+$  dependent on the solution pH as shown in Eq. 1 (Lorne et al., 1999).



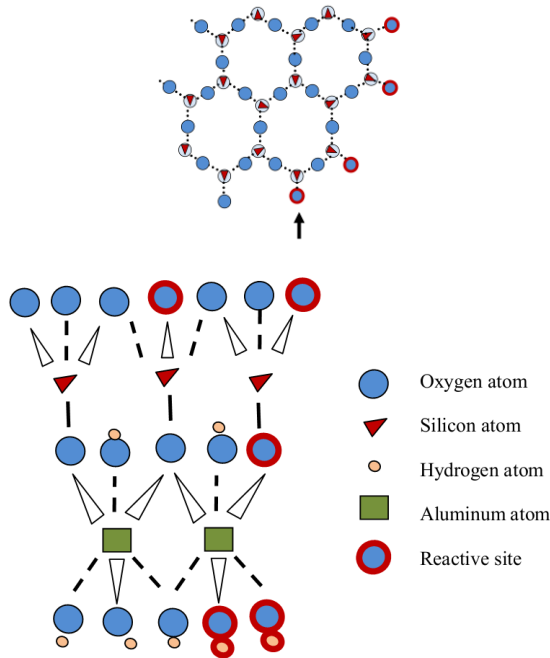
When the solution contains additional cations,  $\text{M}^+$  or anions,  $\text{A}^-$ , these may react with the surface sites as shown in Eq. 2 (Lorne et al. 1999).



The overall surface charge is determined by the number of positive and negative adsorbed ions. The pH at which the surface charge is zero is referred to as the point of zero charge, PZC, (Rudzinski et al., 1999).

The kaolinite mineral ( $\text{Al}_2\text{Si}_2\text{O}_5(\text{OH})_4$ ) consists of a silica sheet bonded to an alumina sheet as illustrated in Figure 2. In the silica sheet, silica tetrahedra are arranged in a hexagonal pattern so that 3 oxygens are shared between tetrahedra, and the remaining oxygens point in the same direction (Figure 2, top). These oxygens are bonded to two aluminum atoms from the alumina sheet (Figure 2, bottom). It consists of alumina octahedra where an aluminum atom is bonded to 4 hydroxyl groups and to two oxygens from the tetrahedral sheet. The hydroxyl groups in the alumina sheet are shared between two octahedra. It has often been assumed only the broken bonds on the edges result in surface sites that interact with water molecules and have a pH dependent surface charge. The faces on the other hand are assumed to

be charged due to ion substitution in the silica and alumina sheets. However, the partially ionic nature of the covalent bonds in the silica tetrahedra and alumina octahedra leaves the surface groups on the faces polarized, which can allow ion adsorption. Data from Huertas et al. (1998), Gan and Franks (2006), and Gupta and Miller (2010), show that the face charges are affected by the pH which indicates ion adsorption to surface sites. Sites associated with Al atoms have a different reaction constant than those associated to Si atoms, and those associated to broken bonds will differ from those on the faces. Therefore a distinct surface charge is expected on the two faces and on the edges of the kaolinite.



**Figure 2:** Representation of the kaolinite structure, Top: 001 face of the tetrahedral silica sheet; silica tetrahedra form a hexagonal pattern with oxygens on the tips of the tetrahedra all pointing below the plane (represented by light blue circles). Bottom: side view of kaolinite mineral indicated by arrow. Oxygen atoms below the silicium atoms are shared with aluminum atoms in the octahedral sheet. These are bonded to hydroxyl groups.

Different methods exist to approximate the surface charge,  $\Psi$ . Potentiometric titrations are used to determine the number of protons adsorbed to the mineral surface. Electro-kinetic methods are used to determine the charge on the interface between immobilized ions and mobile ions in the EDL, the zeta potential,  $\zeta$ . The location in the EDL of this interface is a matter of debate (Elimelech et al., 1995), and due to shielding by adsorbed ions the magnitude of  $\zeta$  will be smaller than the surface

potential. Experimentally, the absolute magnitude of  $\zeta$  is found to decline with increasing ionic strength (Adekola et al. 2010). However,  $\zeta$  is often used to approximate  $\Psi$  due to a lack of alternative data.

For kaolinite particles, these methods yield a charge that is averaged over the entire particle. Atomic force microscopy (AFM) allows the determination of the surface charge on the individual faces of the kaolinite mineral. The interaction force between a surface and a probe with a known surface charge is measured, and DLVO theory is used to derive the surface charge of the mineral. Gupta and Miller (2010) measure the charge on the alumina and silica face of well crystallized kaolinite. To determine the edge charge, Gupta et al. (2011) compare the face surface charges from AFM data to data from potentiometric titration. At pH 6, this results in a surface charge density of the edges that is one order of magnitude greater than that on the faces. The relative contribution of the faces and edges to the average surface charge of the particle depends on the ratio of the edge to face surface areas.

PZC data at ambient temperature for quartz and kaolinite from different studies is shown in Table 1.

**Table 1:** PZC values of quartz surface and average on kaolinite particle from Kosmulski (2006). Data for alumina and silica faces and edges of kaolinite from Gupta et al. (2011).

quartz	kaolinite average	alumina face	silica face	kaolinite edges
<3	<2.4 - 6	6-8	<4	4-6

The PZC for the average charge on kaolinite particles shows significant variation. In addition to the size and shape of the particles, ion substitution and the degree of crystallinity affect the relative numbers of face and edge surface sites. Du et al. (2010) observe stepped faces on kaolinite with a poor crystallinity, increasing the relative number of broken bonds and reactive sites.

### Effects of temperature on surface charge

The temperature affects the equilibrium constants for the reactions on the surface sites (Brady et al. 1996), and thereby the surface charge. The effect of temperature on the surface potential,  $\Psi$ , resulting from protonation of an oxide surface has been estimated using Eq. 3 by Rudzinski (1999) and by Schembre and Kovscek (2005).  $e$  is the elementary charge,  $k_B$  is the Boltzmann constant and  $T$  the temperature (K).

$$\Psi = 2.303 \frac{k_B T}{e} (PZC - pH) \quad \text{Eq. 3}$$

Eq. 3 is based on the Nernst law for the surface charge in the presence of potential determining ions.

The presence of other ions that interact with the surface sites can affect both the surface charge and the effect of temperature on this. The change of the  $\Psi$  as a function of  $T$  is greater when the difference between the PZC and the solution pH is greater, hence injection of fluid with a pH of 6 or higher can be expected to cause a stronger reduction of the  $\Psi$  for quartz than for kaolinite.

Based on a review of published experimental data Kosmulski (2003) suggests that the PZC is reduced when the temperature is raised. Considering that the PZC for the alumina face of kaolinite reported by Gupta and Miller (2010) is between pH 6 and 8, heating in a solution of pH 6 may result in the  $\Psi$  switching from positive to negative. The magnitude of the  $\Psi$  of the alumina face will change little due to the small value of ( $PZC-pH$ ).

There are relatively few experimental studies addressing the effect of temperature on the surface charge of quartz and kaolinite. Figure 3 shows streaming potential data for the  $\zeta$  of quartz and kaolinite in solutions with a pH 4 or higher. Both minerals have an average negative  $\zeta$  in this pH range, and heating causes a reduction of the  $\zeta$ . The differences between the measured values can be attributed to the differences between the ionic strengths and ionic species in the solutions used. A difference between the crystallinity of the samples tested, and hence the surface site density, can also contribute to the difference in the data. Both datasets show the  $\zeta$  of quartz is reduced to a greater extent by heating at pH 8 than at pH 6, as predicted by Eq. 3.

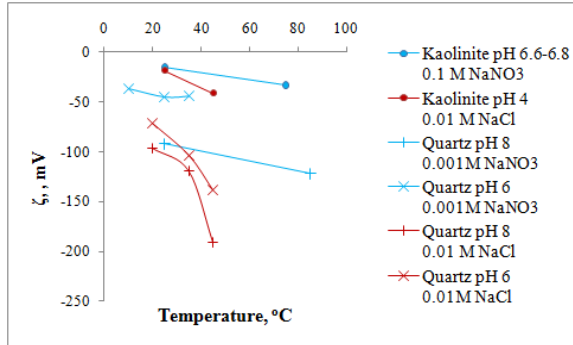


Figure 3: Data of  $\zeta$  as function of temperature for kaolinite and quartz. Red data Rodriguez and Araujo (2006); blue data Ramachandran and Somasundaran (1986).

The effect of heating on the surface potential in a solution with pH 6 is estimated in Figure 4. This shows  $\zeta$  data for quartz and kaolinite and  $\Psi$  data for the two sides of kaolinite and gibbsite (a mineral with the same structure as the alumina sheet), as well as the temperature dependency of these estimated from Figure 3 and Eq. 3. Due to the small difference between the PZC of alumina and the pH, the  $\Psi$  is

expected to change little as a consequence of heating. The  $\Psi$  of the kaolinite silica face is in the same range as the  $\zeta$  of the quartz mineral surface. The  $\zeta$  data may under-estimate the  $\Psi$  due shielding by adsorbed ions, however, the lack of alternative data justifies the use of the  $\zeta$  for quartz.

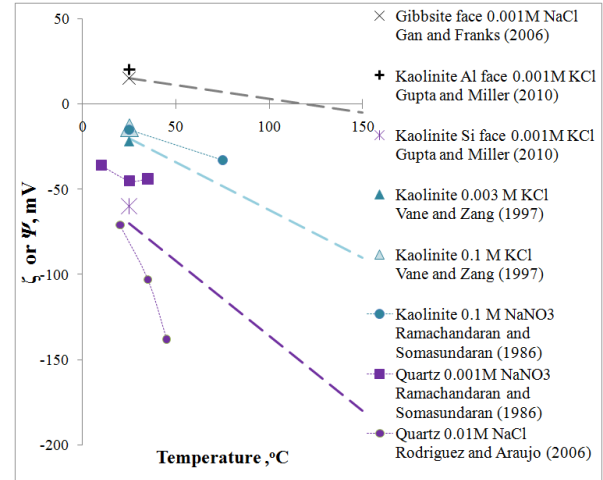


Figure 4: Data of  $\Psi$  (crosses) and  $\zeta$  (closed symbols) and estimated  $\Psi$  as a function of temperature for: Alumina face of kaolinite (gray); average for kaolinite particle (blue) and the quartz mineral surface (purple).

## DLVO INTERACTION ENERGY

The DLVO theory is used to predict the interaction energy between the quartz grains and the kaolinite particles. The interaction energy results from the summation of the EDL interaction and the van der Waals interaction energies (Eq. 4).

$$\Phi_T = \Phi_{vdW} + \Phi_{EDL} \quad \text{Eq. 4}$$

The van der Waals interaction energy,  $\Phi_{vdW}$ , results from spontaneous magnetic and electrical polarization due to the proximity of particles and acts over a short separation distance. For the interaction between a flat plate and a sphere of radius  $a_1$  separated at a distance  $h$  this is calculated by Eq. 5 (Bedrikovetsky et al. 2010).

$$\Phi_{vdW} = \frac{A_{132}}{6h} \left[ \frac{2 * \left(1 + \frac{a_1}{h}\right)}{\frac{a_1}{h} * \left(2 + \frac{a_1}{h}\right)} + \ln \left( \frac{\frac{a_1}{h}}{2 + \frac{a_1}{h}} \right) \right] \quad \text{Eq. 5}$$

$A_{123}$  is the Hamaker constant for the interaction between two different materials that have Hamaker constants  $A_{11}$  and  $A_{22}$  in a medium with Hamaker constant  $A_{33}$  as given in Eq. 6 (Visser, 1972).

$$A_{132} = (\sqrt{A_{11}} - \sqrt{A_{33}})(\sqrt{A_{22}} - \sqrt{A_{33}}) \quad \text{Eq. 6}$$

Values of  $8.86 \times 10^{-20}$  J for silica,  $6.5 \times 10^{-20}$  J for quartz  $1.52 \times 10^{-19}$  J for alumina and  $3.70 \times 10^{-20}$  J for water from Bergström (1997) are used. The Hamaker constant for a kaolinite particle is approximated as the geometric mean of the values for silica and alumina ( $1.16 \times 10^{-19}$  J). These values are assumed to be constant with temperature as a first approximation.

The surface charge on the minerals attracts ions with an opposite charge forming a diffuse layer on the particle surface. The Poisson-Boltzmann relation gives the variation of electrical potential with distance from the surface in the EDL, subject to the assumption that the surface charge is uniformly distributed on the mineral surface, the ions are point charges and the properties of the fluid are constant with distance from the surface (Elimelech et al., 1995). The interaction energy,  $\Phi_{EDL}$ , due to the potential in the EDL on a quartz grain and that on a kaolinite particle is approximated by Eq. 7 (Elimelech et al., 1995) for a spherical particle and a flat surface,

$$\Phi_{EDL} = \frac{\epsilon_0 \epsilon_{rs} a_i}{4} \left( \begin{aligned} &2\Psi_1 \Psi_2 \ln \left( \frac{1 + \exp(-\kappa h)}{1 - \exp(-\kappa h)} \right) \\ &+ (\Psi_1^2 + \Psi_2^2) \ln(1 - \exp(-2\kappa h)) \end{aligned} \right) \quad \text{Eq. 7}$$

where  $\epsilon_{rs}$  is the relative permittivity of the solution  $\epsilon_0$  is the vacuum permittivity, and  $\Psi_1$  and  $\Psi_2$  are the surface charges on the quartz surface and the kaolinite respectively. The inverse Debye parameter,  $\kappa^{-1}$ , characterizes the distance over which the surface charge is reduced to a value  $1/e$  i.e.  $1/2.72$  (Lyklema, 1995). Eq. 8 expresses  $\kappa^{-1}$  where  $I$  is the ionic strength and  $N_A$  is Avogadro's number.

$$\kappa^{-1} = \left[ \frac{\epsilon_{rs} \epsilon_0 k_B T}{N_A e^2 I} \right]^{1/2} \quad \text{Eq. 8}$$

It assumes that the surface potential on the kaolinite particles is uniformly distributed.

The total interaction energy,  $\Phi_T$ , between a kaolinite particle and the quartz surface is computed using the surface potentials given in Table 2. The values for kaolinite represent the average charge on the kaolinite particles. Figure 5 shows the interaction energy between quartz and kaolinite scaled by  $k_B T$ , a

measure of the thermal energy. Negative interaction energy represents attraction.

Table 2:  $\Psi$  values estimated from data in Figures 3 and 4. Values at  $150^\circ\text{C}$  have low accuracy due to extrapolation beyond the data range.

	$\Psi$ , mV ( $25^\circ\text{C}$ )	$\Psi$ , mV ( $150^\circ\text{C}$ )
<b>Quartz grain</b>	-70	-180
<b>Kaolinite average</b>	-10	-80

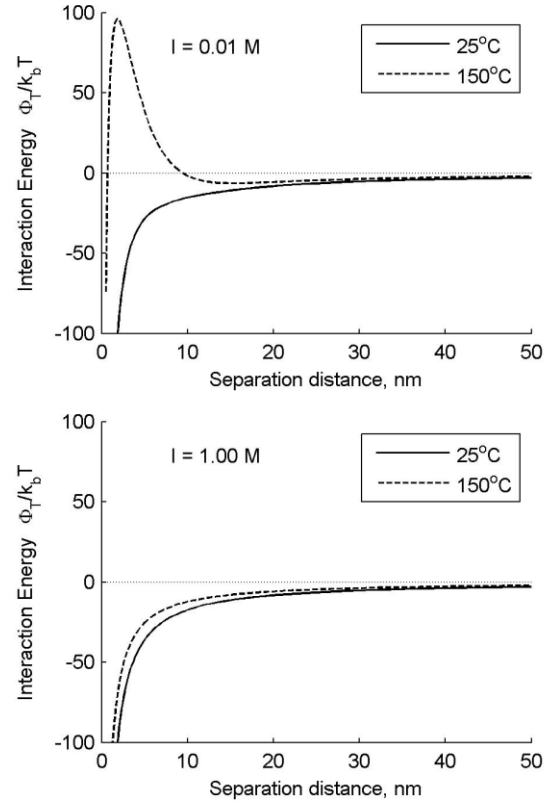


Figure 5: Interaction energy,  $\Phi_T$ , between quartz surface and a spherical kaolinite particle ( $a_i = 0.5 \mu\text{m}$ ).

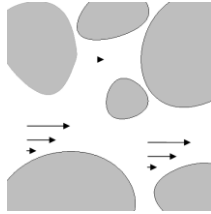
At  $25^\circ\text{C}$  the attractive forces between the quartz surface and the particles with the average kaolinite surface potential are dominant. Heating up to  $150^\circ\text{C}$  changes the interaction energy into repulsion in the solutions with an ionic strength of  $0.01 \text{ M}$ . For separations smaller than  $1 \text{ nm}$  the van der Waals forces dominate. However, the fluid layer adsorbed to the mineral surface can be expected to prevent the particle from approaching the surface this closely (Khilar and Folger, 1984). The interaction energy at  $150^\circ\text{C}$  shows a secondary energy minimum around  $12 \text{ nm}$  separation. It is suggested that particles may be captured via the minimum provided that the interaction energy is sufficiently attractive (Redman et al., 2004). Hydrodynamic forces can be expected

to be more significant than DLVO forces at the distance.

When the ionic strength of the solution is 1 M, the interaction energy between the quartz and the kaolinite particles is negative at both 25°C and 150°C. The high ionic strength reduces the range of the EDL force so that van der Waals attraction dominates.

### **HYDRODYNAMIC FORCES**

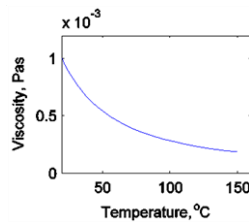
The hydrodynamic force that the fluid exerts on a particle depends on both the fluid velocity and the fluid viscosity. The flow velocity field in pores is non-uniform. Assuming laminar flow and a non slip condition on the solid surface, the flow velocity can be expected to increase with the distance from the quartz grains towards the center of the pores. Additionally, the pore geometry can be expected to result in flow channels as well as in pockets with low flow velocities (Bear, 1972). A schematic representation of expected flow velocity variations is shown in Figure 6.



*Figure 6: Schematic representation of flow from left to right through a sandstone sample. The arrow length represents fluid velocity. The highest velocities are expected in the center of channels parallel to the flow direction.*

The velocity field in Figure 6 implies the hydrodynamic force on particle aggregates bridging the pore throats is greater than that on particles on the grain surface or in pockets with little flow.

Figure 7, computed using the program provided by Mao (Mao and Duan, 2009), shows the reduction in viscosity of distilled water with temperature. Heating from 25°C to 150°C causes a more than 4 fold reduction in the viscosity.



*Figure 7: The viscosity of distilled water decreases non- linearly as a function of temperature.*

### **DISCUSSION**

The kaolinite mobilisation with increasing temperature can be attributed to an increase in the absolute magnitude of the surface potential of both kaolinite and quartz. Different sides of the kaolinite mineral have a different surface charge. Gupta et al. (2011) show that despite the larger surface area of the faces, the surface potential of the edges is dominant in determining the average surface potential of the kaolinite particle due to the higher charge density on the edges. The average surface potential for kaolinite is used to approximate the EDL interaction energy between quartz grains and the kaolinite particles at 25°C and 150°C. This results in repulsion of the particles from the grain surface in solutions with low ionic strength, while it is insensitive to the temperature at high ionic strength due to increased shielding of the surface potential.

The smaller absolute magnitude of the surface potential on the faces compared to the edges of kaolinite can be expected to influence the interactions between suspended particles. The change in the surface potential of the alumina face due to heating is expected to be smaller than that of the edges or that of the quartz grains. It may cause an attractive total interaction energy between kaolinite particles that have been mobilized due to heating.

The flowing fluid carries and concentrates particles in the pore throats, where they can aggregate and form bridges, resulting in a permeability reduction. A reduction of the temperature reduces the absolute magnitude of the surface charges and the repulsive interaction energy. It tends to make bridged particles more stable. The effect can be offset by the increase in the hydrodynamic force with cooling which has a greater effect in the flow channels than on the grain surfaces. The dislodged particles may be recaptured via attachment on the surface of the sand grains downstream.

The mechanism correlating the temperature change and the permeability change requires the domination of both the electrostatic forces and the hydrodynamic drag on the particles. When the flow rate is low, cooling may not cause a remobilisation of aggregated grains. When it is high, heating may not cause additional mobilisation.

Quantification of the DLVO interaction energies and the hydrodynamic forces is complicated by the number of influential parameters. To the best of our knowledge, the effect of the heterogeneous surface charge distribution of kaolinite on the structure of the EDL is uncertain. The values assumed for the surface charges and the Hamaker constants, particularly at elevated temperature are based on the limited data. The characteristics of the porous medium, including i.e., the pore geometry, the mineral content, the crystallisation and the surface roughness, affect both the interaction energy and the hydrodynamic forces.



The computed forces thus serve only for an indicative comparison of the interaction energies.

The above described mechanism is in agreement with the observations of the permeability reductions in kaolinite bearing sandstones reported by Weinbrandt and Ramey (1975), Somerton and Mathur (1976), Casse and Ramey (1979), Sydansk (1980), Jing et al. (1992), Baudracco and Aoubouazza (1995), and Schembre and Kovscek (2005). The temperature is not subsequently reduced in all studies, but where it is the case the permeability reduction is found to be largely reversible. When the temperature is raised in successive increments the permeability also reduces in steps. Each temperature step increases the EDL repulsive force, provoking fines mobilisation from the grain surface, and reduces the fluid viscosity, reducing the destabilizing hydrodynamic force on the aggregated particles.

In a number of experiments kaolinite mobilisation causes permeability reduction during injection at room temperature (Sydansk, 1980; McKay and Brigham, 1984; Gobran et al., 1987). Heating has a negligible effect in some cases (McKay and Brigham, 1984; Gobran et al., 1987). In the tests by Sydansk (1980), permeability reduction is observed during injection at room temperature and heating causes an additional permeability reduction. The interstitial flow velocity in the first two tests is higher than in a number of tests where reversible permeability reduction occurs as a consequence of heating; however, the number of publications with reported flow velocity is limited. To examine whether the interaction of hydrodynamic and EDL forces is as proposed, experiments where permeability is measured as a function of temperature for different flow velocities are needed.

A high ionic strength may result in the reduction of the thickness of EDL and the repulsive EDL force (Figure 5). DLVO theory is used to explain particle mobilisation caused by a reduction of the ionic strength of the saturating solution in a number of studies, e.g., Khilar and Folger (1984). Experiments by Sydansk (1980) as well as by Schembre and Kovscek (2005) with NaCl show the effect of temperature on permeability is greater in solutions with a lower ionic strength. No permeability reduction is observed in tests with 0.79 M KCl in two different kaolinite bearing sandstones (Wei et al., 1986); on the other hand, tests with a 0.86 M solution of NaCl (Jing et al., 1992), as well as those with 1.0 M NaCl and  $\text{CaCl}_2$  (Baudracco and Aoubouazza, 1995) do show permeability reduction with temperature, which is reversible with cooling in the latter two cases.

Besides the flow velocity differences, there may be other reasons why the temperature increase causes the permeability reduction in some tests with a high ionic strength but not in others. The size and charge of dissolved ions affects the strength of their interaction with the mineral surface (Khilar and

Folger, 1984) and thereby the surface charge and EDL force. The surface charge of other rock forming minerals, such as feldspars, can be expected to differ from that of quartz. The clay content and the distribution of clay in the sandstone pores affect the amount of particle mobilisation and bridging. Based on scanning electron microscopy images, Tchistiakov (2000) suggests that kaolinite particles dispersed throughout the sandstone are mobilized to a greater extent than the kaolinite present in relatively isolated pockets that can result from the weathering of a feldspar grain. Furthermore, the possibility that the permeability change is due to a different mechanism is not ruled out in the tests compared here. Cement dissolution can release particles causing clogging (Blair et al. 1984). Experimental error, in particular contamination by colloidal particles released from the experimental equipment at elevated temperature, is detected in a number of experiments (Potter et al., 1980; Stottlemire, 1980) and not ruled out in other tests.

## **CONCLUSION**

Kaolinite mobilisation is one of the mechanisms that can contribute to the permeability reduction in sandstones as a consequence of heating. The available data indicate that quartz and kaolinite have an average negative surface charge that becomes more negative due to heating. As a result, the repulsive interaction energy between the quartz grains and kaolinite particles is increased. Kaolinite is a sheet mineral consisting of two different sheets. It has a different surface charge on the two different faces as well as on the edges of the mineral. The interactions between different sides of the suspended kaolinite particles may promote particle aggregation and bridging in the pore throats which subsequently reduces the permeability of sandstones. The increase in the permeability at reduced temperatures can be attributed to the enhanced hydrodynamic force on the bridged particles and attachment of particles to the quartz grain surface.

Due to the number of parameters affecting particle mobilisation and permeability reduction, comparison of published data is insufficient to verify the proposed mechanism. Further investigation under controlled conditions is required to verify the proposed mechanism of particle induced permeability reduction.

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